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STATUS REPORT

GROUND DISPOSAL OF HIGH LEVEL RADIOACTIVE WASTES

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Introduction

In October 1953, a preliminary proposal entitled "The Safe and Economical Disposal of Effluents from the Nuclear Power Industry"(1) was prepared by representatives of the ORNL Health Physics Division, and given limited distribution. This "Draft" was followed in May 1954, by "A Revised Proposal to Study Ground Disposal of High Level Radioactive Liquid Wastes,"(2) which presented a more realistic definition of the studies to be pursued at ORNL. Since the preparation of the "Revised Proposal", changes in ORNL program plans (Project 25) required further revision of the research program envisioned in that document.

The purpose of the present report will be to redefine the responsibilities of the Sanitary Engineering Research Section of the Health Physics Division in the disposal of high level reactor wastes and to report the progress made in the past six months.

At this time the Sanitary Engineering Research Section wishes to express its appreciation for being invited to the three conferences on waste disposal sponsored by the Sanitary Engineering Division, AEC, and the Johns Hopkins University. These were the conferences on ground disposal, ocean disposal, and deep well disposal of high level radioactive wastes.

A Statement of the Immediate ORNL Problem

The proposed research program of the Sanitary Engineering Section is concerned primarily with the release of high level radioactive wastes into the weathered overburden and the storage of these materials in shallow earthen pits. Before release of these wastes into the overburden can be justified, it is necessary to obtain information on the exchange capacity of the soil, the natural exchange complex, the interactions between the waste solution and the soils, the chemical pollution effects, and the rate of movement and direction of flow. In the case of containment in shallow earthen pits, knowledge is required relative to the effect of mineral and bituminous barriers, the external hazards resulting from such storage, and the effect of the heat of decay on the storage of the wastes. All of this information is needed to prepare engineering criteria for the design of prototype disposal pits.

As pointed out in the "Revised Proposal",[2] this program may involve the handling and possible disposal of 10^9 curies of fission products per year. An estimate of the fission product yield of a typical homogenous reactor was also given and will not be repeated. It will be recalled, however, that the nuclide concentrations in the processing plant effluent and in an assumed pit at equilibrium were determined. The total activity in the processing plant effluent amounted to 836,500 curies per day and in the pit, after 10 years use, 6.25×10^8 curies.

Furthermore, using the same distribution of fission products a preliminary estimate was made of the temperature rise in the pit as a function of radioactive decay and it was found that sufficient heat is available to melt the surrounding medium. In other words, sufficient heat is available to fuse and permanently fix the radioactive materials in place. The large quantities of radioactivity and the high temperatures are associated with the discharge of relatively small volumes of waste, approximately 1500 gals per day.

In the "Revised Proposal" the hazard factor represented by each nuclide was determined, based upon its fission yield and on the MPC values in air and water recommended by the NCRP and the ICRP. These estimates showed that the most hazardous radioisotope was Sr^{90} both in terms of ingestion and inhalation. In the case of ingestion, for example, the most critical isotopes following Sr^{90} were Sr^{89}, I^{131}, Nb^{95}, Ba^{140}, Ce^{144} - Pr^{144}, Y^{91}, Cs^{137} - Ba^{137}, Y^{90}, Zr^{95}, Ru^{103}, and Ru^{106} - Rh^{106} for the chemical processing plant, and for the pit after 10 years use the order of hazard after Sr^{90} was Nb^{95}, Sr^{89}, Cs^{137} - Ba^{137}, Ba^{140}, Y^{90}, Zr^{95}, Ru^{103}, and Ru^{106} - Rh^{106}.

The critical nuclides defined above have been represented in waste solutions studied in the laboratory and on a pilot plant scale by six radioisotopes, namely: Sr^{90}, Y^{90}, Zr^{95}, Nb^{95}, Cs^{137} - Ba^{137}, and Ru^{106} - Rh^{106}. A 1.6M $Al(NO_3)_3$ solution containing 0.2 M HNO_3 and sufficient added stable isotopes to represent the proper concentration of fission products served as the stock solution in all investigations of simulated high level wastes, and each of the radioisotopes was added as a tracer to note the effect of a specific treatment process on that isotope. This solution was representative of a waste from the chemical processing of solid fuel elements.

Program of Investigations

The waste research program at ORNL includes an evaluation of the suitability of ground disposal for ART and HRT waste effluents. Geologic, pedologic, hydrologic, meteorologic, and ecologic parameters are under consideration. As in the past, other agencies are participating in these studies with the Health Physics Division at ORNL. Included thus far are representatives of the U. S. Public Health Service, U. S. Geological Survey, U. S. Weather Bureau, Tennessee Valley Authority, and the Engineers Research and Development Laboratories, U. S. Army. The U. S. Public Health Service has been encouraged to accept broader responsibilities and provide increased technical assistance in the execution of this program. Technical and financial support for the study of the engineering problems associated with ground disposal of high level wastes from nuclear power reactors will be provided by the Division of Engineering, AEC, and for the study of the hazards associated with the disposal of radioactive wastes from reactors by the Division of Biology and Medicine, AEC.

In the sections that follow, a description is given of the continued program of research and the progress that has been made since submittal of the "Revised Proposal".[2] It is presented in three parts: air pollution, liquid waste disposal, and ceramics.

Air Pollution

The use of large pits filled with sand and gravel has been suggested for the disposal of high level wastes. Liquid wastes introduced into the bottom of the pit concentrate by evaporation. Sand is provided to de-entrain aerosols and to serve as a radiation shield.

Since little or no information exists on the effectiveness of sand for filtering aerosols at the low boil up velocities expected in pits, the main purpose of this study will be to investigate the efficiency of sand layers for aerosol filtration at low air velocities. Some laboratory scale experiments of aerosol entrainment, using tracer amounts of activity, have been carried out[3], pending installation of a steel "aerosol entrainment well" 20 ft deep and 3 ft in diameter in the field. This aerosol entrainment well is shown in Figure 1. The laboratory studies, made with boiling alkaline radioactive solutions, indicated decontamination factors of about 10^6 due to distillation alone[3]. The field experiments are designed to determine the thickness of sand required to obtain decontamination factors of the order of 10^{12} from boiling radioactive solutions. Other laboratory studies of the efficiency of sand filters at low flow velocities showed that maximum penetration occurred with 0.5 μ diameter aerosol particles[3]. A rapid method of particle size determination was developed and involves the determination of aerosol penetration through a lead-shot column as a function of particle

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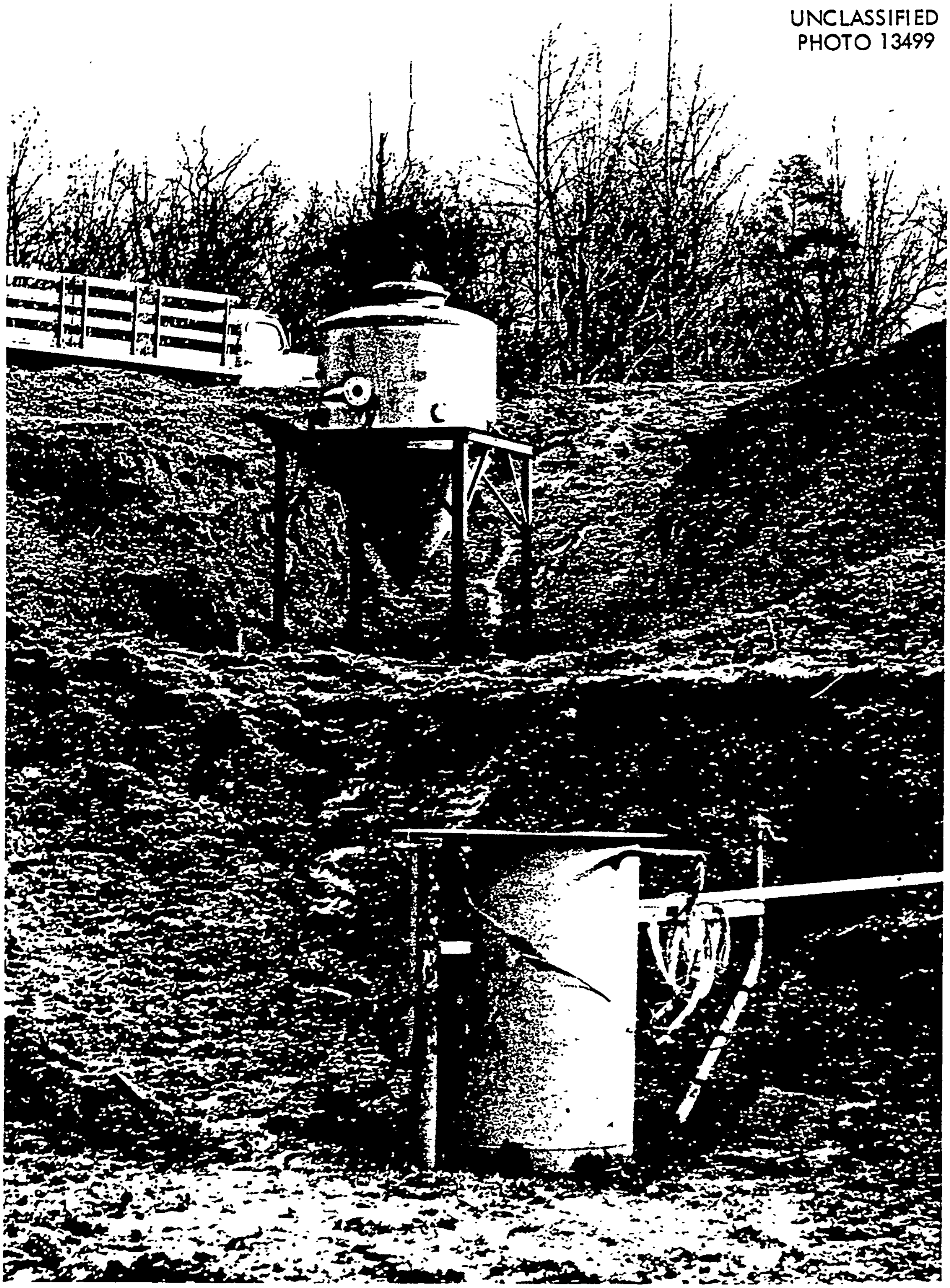

Fig. 1. Experimental Aerosol Entrainment Well in Foreground, Dissolver Tank in Background.

size in the 0.1 to 1.0 μ radius range. Approximately two minutes are required for the determination of the particle size. A typical penetration curve is shown in Figure 2.

For the study of the dispersal of airborne contamination from stacks and pit sources, a smoke generating unit has been developed. A tracer element will be injected into the base of the stack, and ground concentration will be determined following collection and activation analyses of the air samples. The use of the smoke generator is shown in Figure 3. Meteorologists from the U. S. Weather Bureau office at Oak Ridge will assist the ORNL research group in these studies.

A cloud chamber, shown in Figure 4, was developed for counting sub-micron particles [4], and, when used in conjunction with the diffusion tube,[5] particles approximately 0.01 μ radius were measured. By air dilution experiments, the cloud chamber was proved to be linear with respect to particle concentration. It was also found that the diffusion battery[5] method of aerosol particle size determination is more accurate when experimental results are extrapolated to zero air flow through the battery. A schematic diagram of the diffusion battery is shown in Figure 5.

Liquid Waste Disposal

Activities in this field fall into three broad classifications: wastes associated with nuclear power industry, ORNL process wastes, and correlated activities.

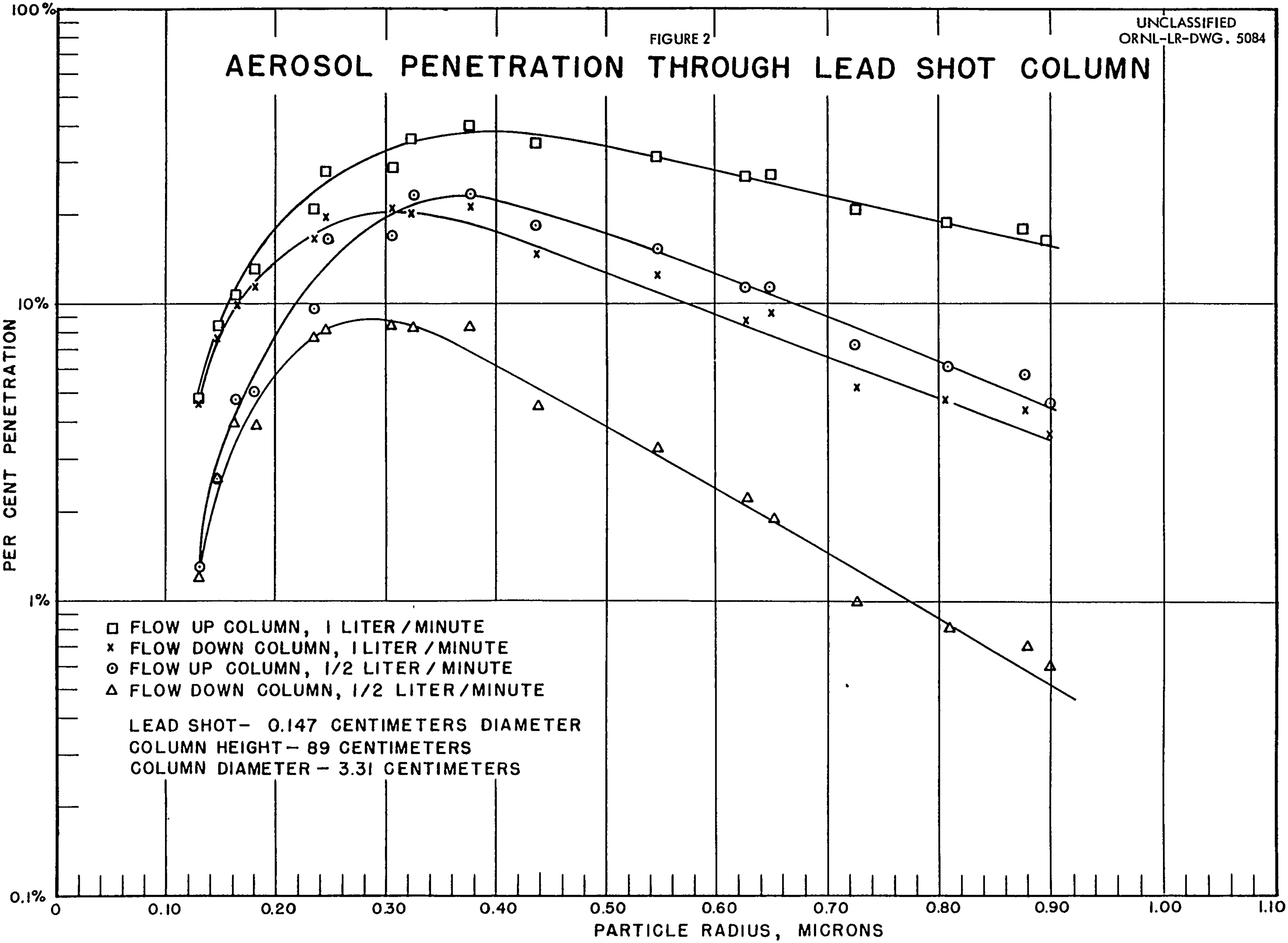

FIGURE 2

AEROSOL PENETRATION THROUGH LEAD SHOT COLUMN

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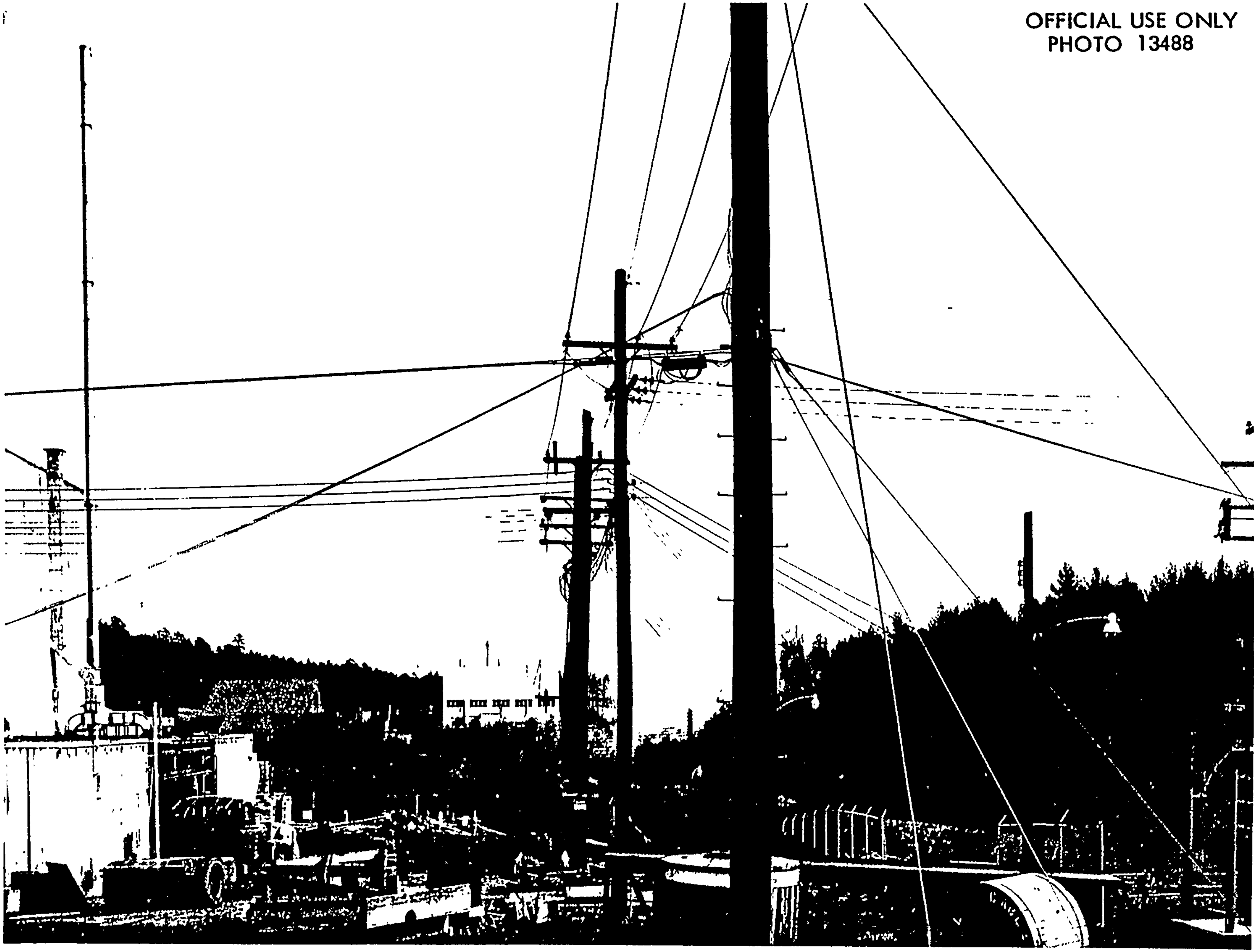

Fig. 3. Smoke Generator in Use. Note Deposition of Smoke in Middle Foreground Between Poles.

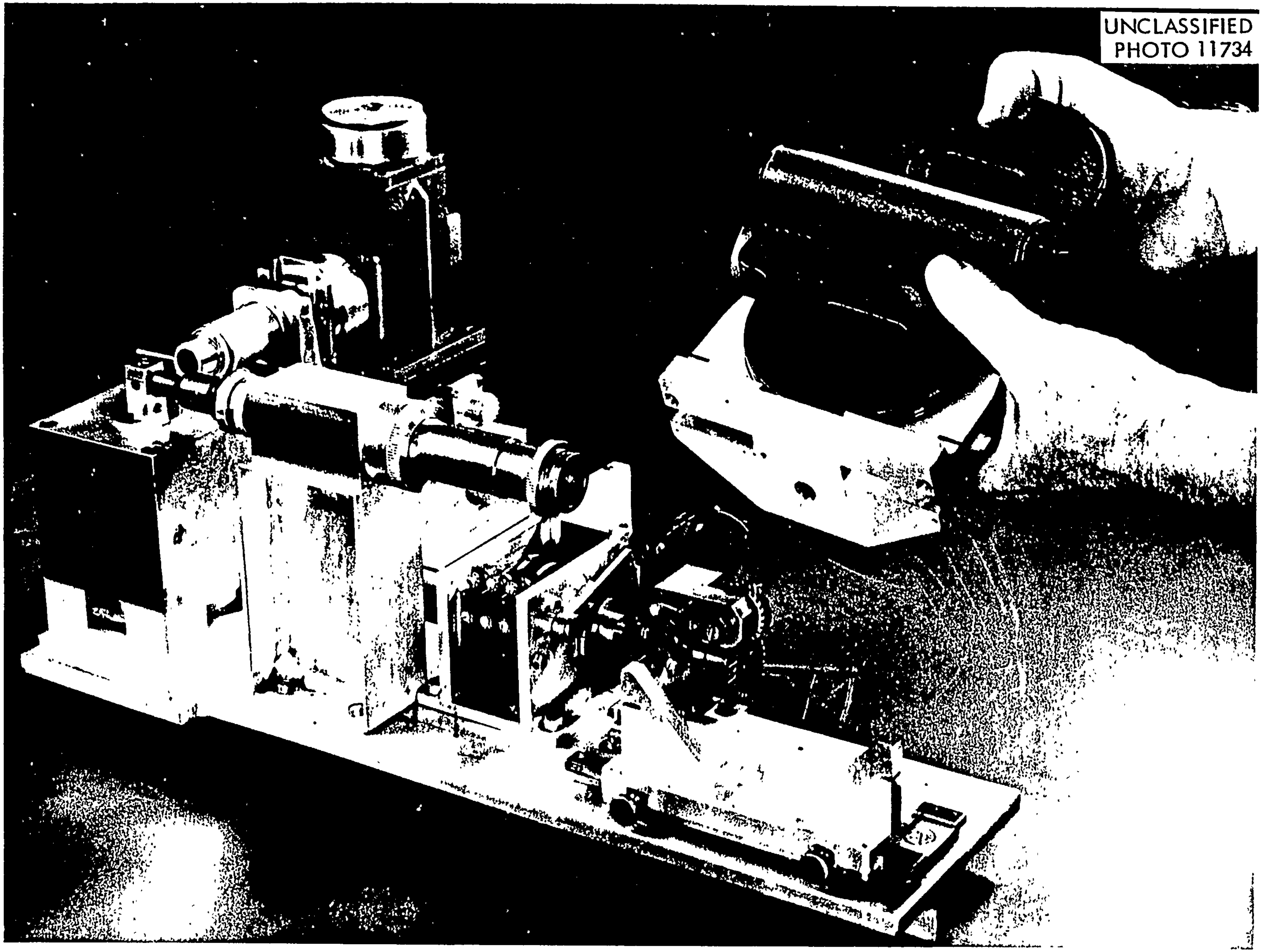

Fig. 4. Cloud Chamber.

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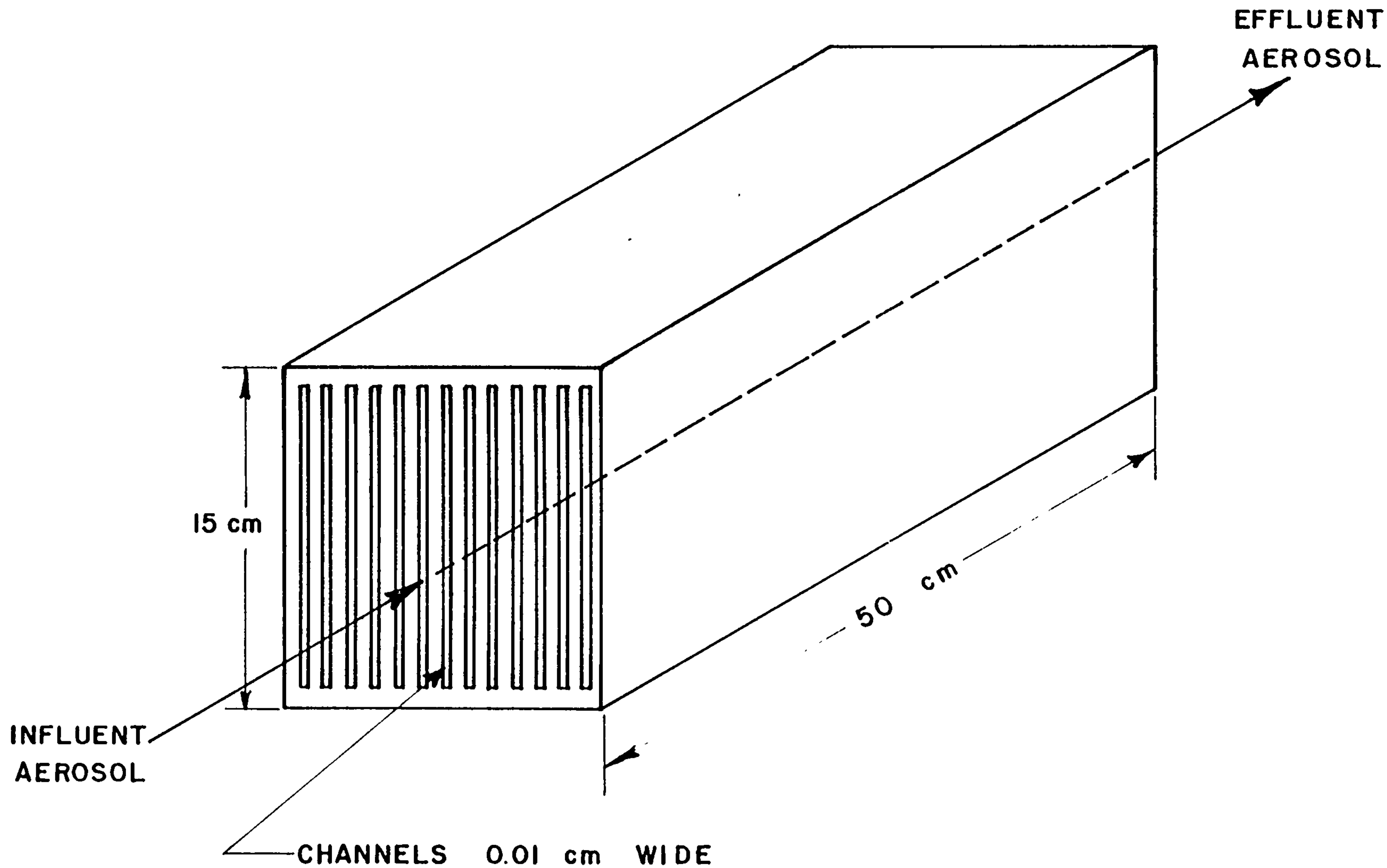

Fig. 5. Diffusion Battery.

Wastes Associated with Nuclear Power Reactors. These studies may be defined as relating chemical and engineering laboratory findings of the interactions between the waste solutions and the soil formations to field applications where the influence of geology and hydrology must be recognized and evaluated.

The chemical and engineering laboratory investigations were concerned with the composition of waste solutions, the properties of soils in the ORNL area, the characteristics of ground water flow and movement, the migration of radioactive and chemical constituents of the waste through the soil, the determination of the exchange and adsorptive capacity of the soil, and the effect of chemical composition of both soil and waste materials on retention and fixation. Consideration also was given to the thermal problem and the possible fixation of radioactive materials in the soils by the inherent heat of decay. The self-sealing tendencies of the wastes in contact with local soils have been investigated and some study has been given to the use of mineral and bituminous pit liners for additional protection.

Because of the high cost of the large amount of NaOH required to neutralize the acid aluminum nitrate dissolver solution containing the fission products, chemical precipitation techniques were evaluated for the removal of the six critical nuclides cited earlier from this acid waste solution. There is a two-fold purpose in these studies: 1) to separate out the critical nuclides (fission products) without precipitating the aluminum, and 2) to reduce the amount of activity discharged to waste pits, thus minimizing the problems associated with the heat generated during decay. These studies will be continued using ion exchange and solvent extraction procedures for separating the fission products from the aluminum solution.

Procedures were developed for the recovery and identification of the critical radioisotopes fixed to soils (Sr, Y, Zr, Nb, Cs, and Ru). Methods were devised for the determination of radioactive strontium and barium in natural waters, sewage and sludges. Concentrations of radiostrontium and barium of $\sim 4 \times 10^{-8}$ and $\sim 10^{-7}$ µc/ml, respectively were found.[6] Studies also showed that ion exchange was the mechanism of removal of cesium on cellulose[7]. The results of this study may assist in defining the nature of the removal of cesium by soil.

The movement of radioactive materials through soil columns was investigated. Tests completed thus far with neutral solutions show that, with the exception of Cs, the order of removal followed that predicted by the Hofmeister lyotropic series[8].

The base exchange capacity, the natural exchange complex, and the permeability of various soils in the ORNL area have been defined both with laboratory soil columns (Figure 6) and for various soils <u>in situ.</u> Specific clay liner materials have been investigated in the laboratory and on a semi-pilot plant scale. Permeability results to date show k_{20} (permeability coefficient at 20° C) values for Tennessee ball clay of 10^{-8} to 10^{-9} cm/sec. Similar studies with asphalt liner materials supplied by the Chemical Technology Division indicate k_{20} values of about 10^{-11} cm/sec.

A 3-ft clay strip liner has been placed in the third ORNL chemical waste pit (see Figure 7) for full scale evaluation under field conditions.

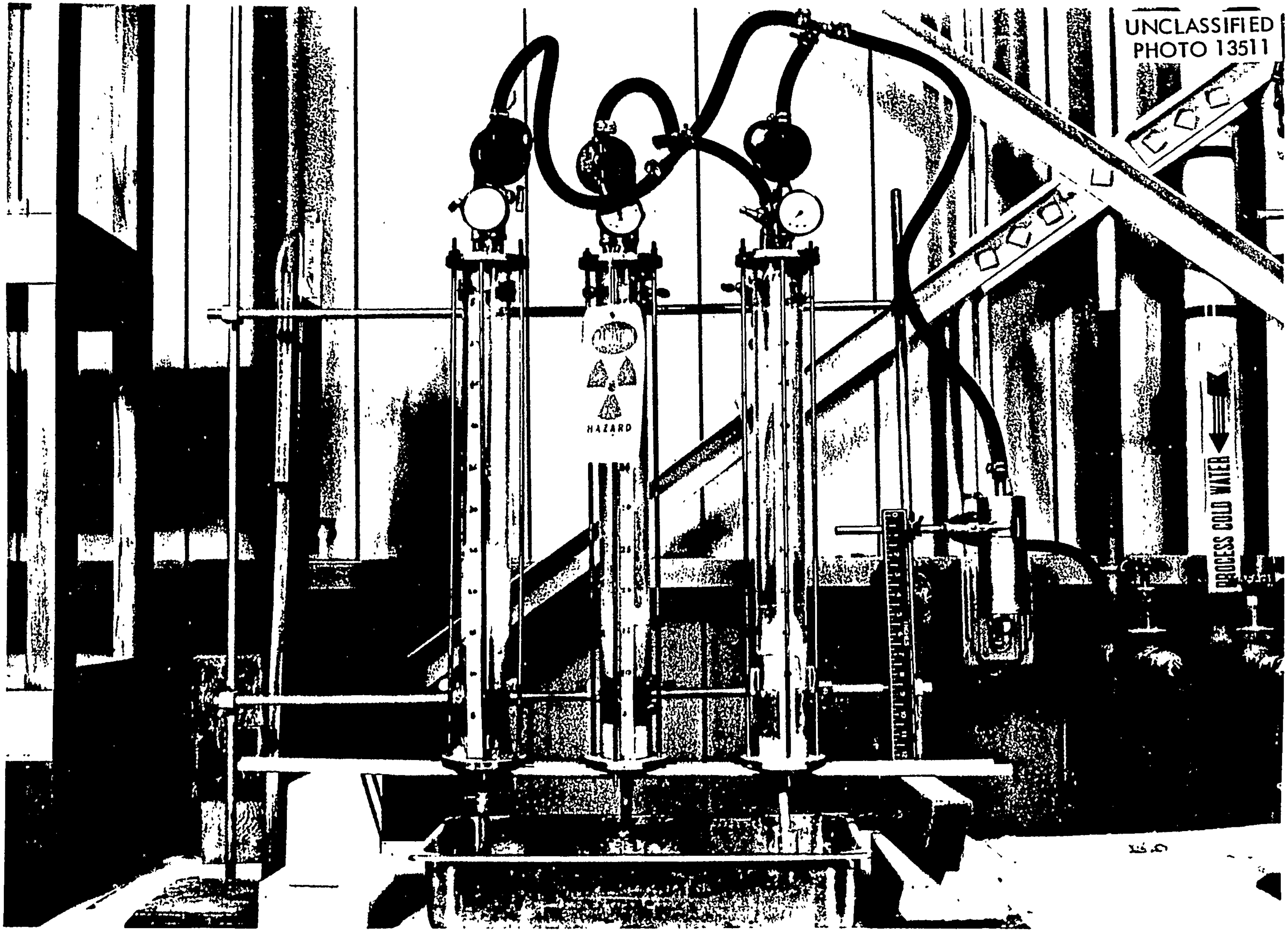

Fig. 6. Laboratory Percolation Columns for Evaluating Permeability of Mineral Liner Materials.

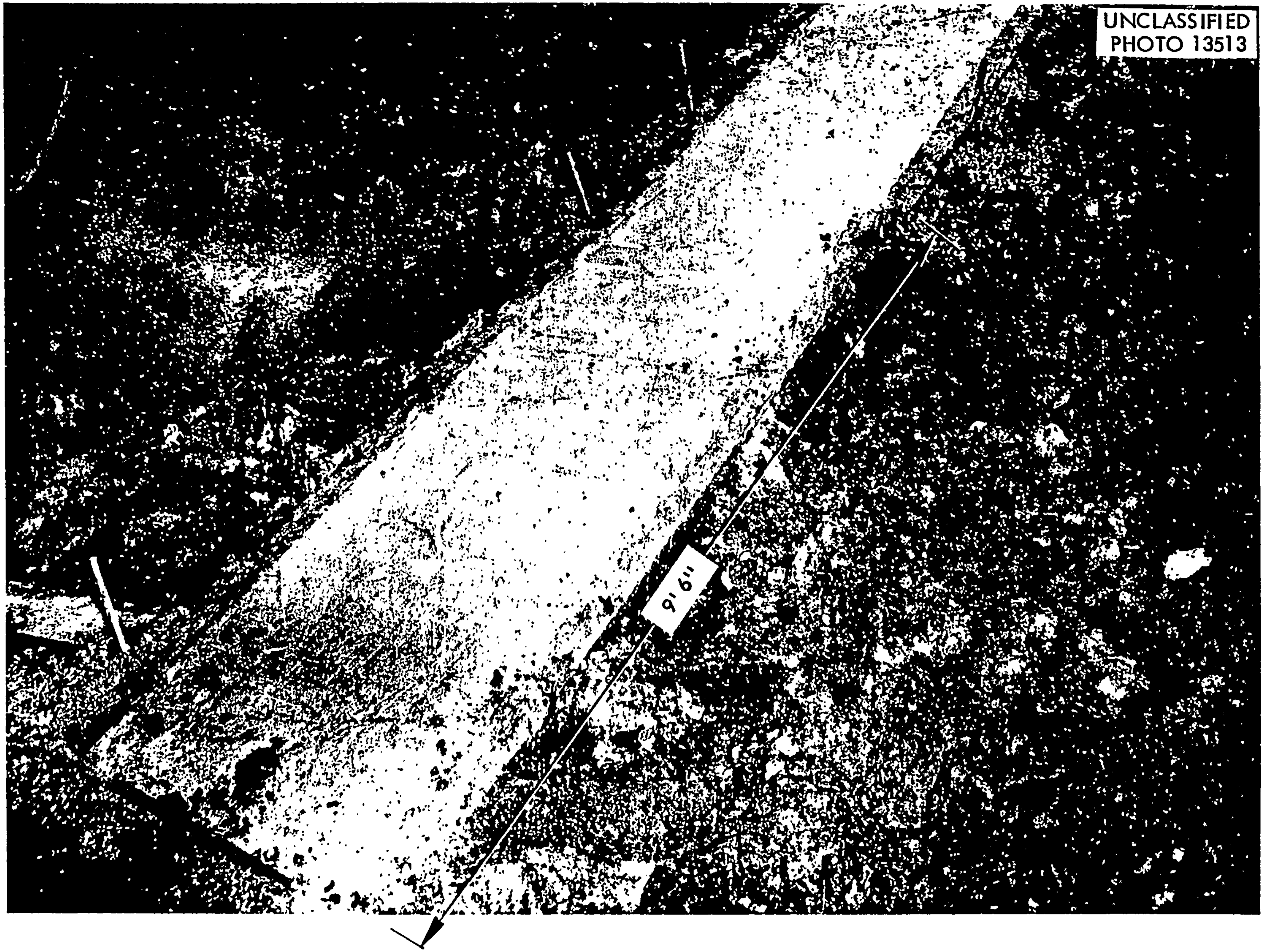

Fig. 7. Three-foot Strip Liner of Tennessee Ball Clay in Place on Side Slope of ORNL Chemical Wast Pit #3.

An asphalt membrane and one ball clay liner over sand in drums are under investigation in this same chemical waste pit.

Model studies are planned to determine better methods of waste distribution to obtain optimum contact between the soil mass and the waste solution to provide maximum retention and fixation of the radioactive constituents.

Field investigations included the study of the movement of ground waters and waste solutions through soil and rock formations in the ORNL area, and have provided data for the location of observation and sampling wells. These data, obtained in the four-acre tract following pressure, pumping, and recharge tests and water level measurements, will assist in the development of criteria for the design and operation of a full-scale field installation.

Two experimental pits, one lined and the other unlined, each approximately 30 ft in diameter and 5 ft deep, were studied. The unlined pit received 741 gallons of a simulated acid dissolver solution on June 9, 1954, from the dissolution of non-irradiated slugs, containing 3.4 gms/liter of uranium and 343.7 gms/liter of nitrates. The movement of chemical pollution was followed by field nitrate tests (diphenylamine spot plate method) and quantitative nitrate and uranium analyses on samples obtained from the observation and sampling wells. A plot of the pit and the direction of travel of nitrates and uranium is shown in Figure 8A. Figures 8B and 8C show water table configurations on May 19, and June 2, 1954, respectively.

The water-tightness of the field applied asphalt tamped clay liner in the experimental lined pit was evaluated by an evaporation study[9].

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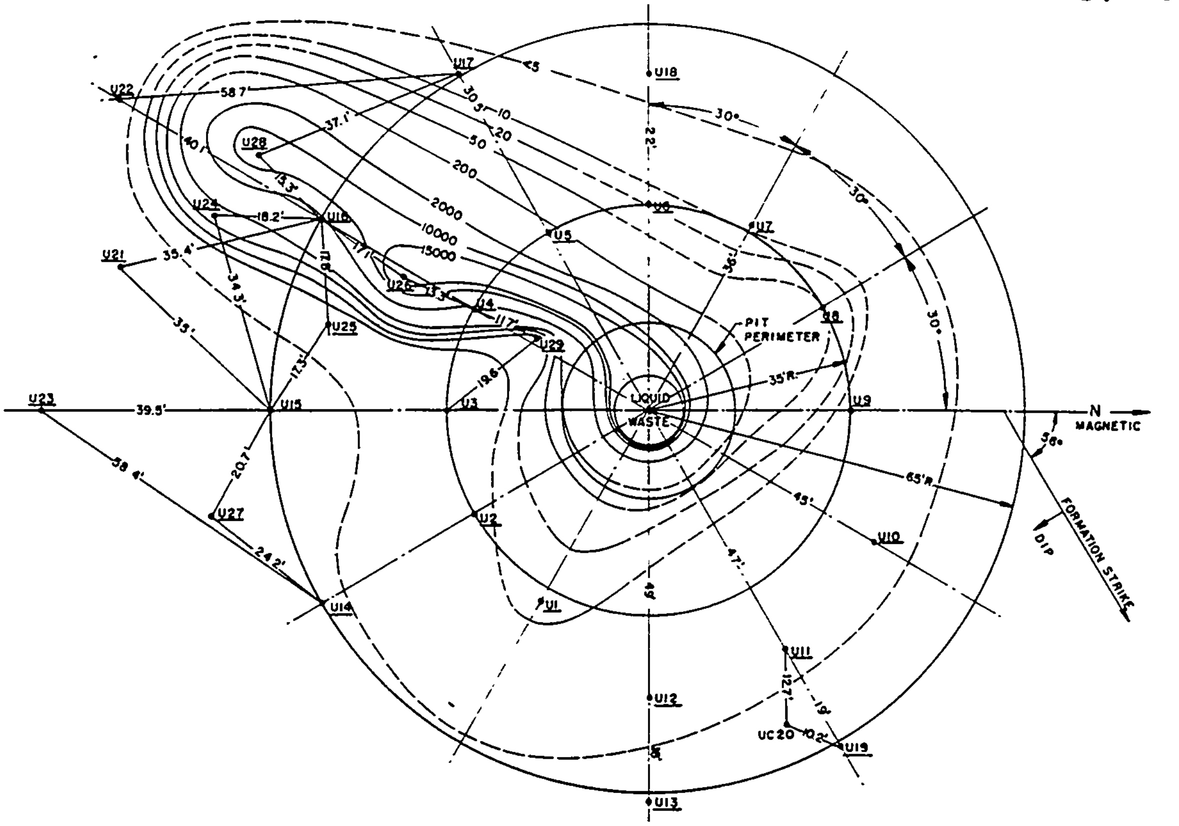

LEGEND

• OBSERVATION AND SAMPLING WELLS (RESULTS OF 9/13-14/54)

50 NITRATE CONTAMINATION CONTOURS IN PARTS PER MILLION BASED ON FIELD AND LABORATORY ANALYSIS—LOCATION APPROXIMATE.

NITRATE CONTAMINATION CONTOURS-LOCATION INFERRED OR PROJECTED.

NITRATE AND URANIUM ANALYSIS
(ALL RESULTS IN PARTS PER MILLION)

SAMPLING DATE / WELL	5/21/54 LABORATORY NITRATES	5/21/54 LABORATORY URANIUM	9/14/54 LABORATORY NITRATES	9/14/54 LABORATORY URANIUM	9/13/54 FIELD NITRATE
U 1	0.1	1.8	12.0	1.0	10+
U 2	0.2	1.1			DRY
U 3	0.1	1.8			+
U 4	8.0	0.5	1600	0.6	10+
U 5	8.4	1.4	214	0.7.	10+
U 6	0.6	0.9			DRY
U 7	0 5	0.8			+
U 8	0.2	0.8			DRY
U 9	0.3	1.4			DRY
U 10	0.5	1.4			DRY
U 11	7 0	0 9			+
U 12	<0.1	0 4			+
U 13	0.4	0.4			+
U 14	0.2	0.9			+
U 15	0.7	0.9			+
U 16	28 0	0.4	1990	9.0	10+
U 17	0.1	0.4			+
U18	0.2	0.9			+
U19	0.3	0.9			+
U20	0.2	1 8			+
U21			<1.0	1 1	5-
U22			1.0	1 0	5-
U23			<1.0	1.3	5-
U24			55.0	0.5	10+
U25			<1.0	0.8	5-
U26			19210	31.0	10+
U27			1 0	2.0	5-
U28			13000	2.3	10+
U29			17 0	0 6	10+

Fig. 8A. Unlined Experimental Pit. Chemical Pollution Flow Pattern.

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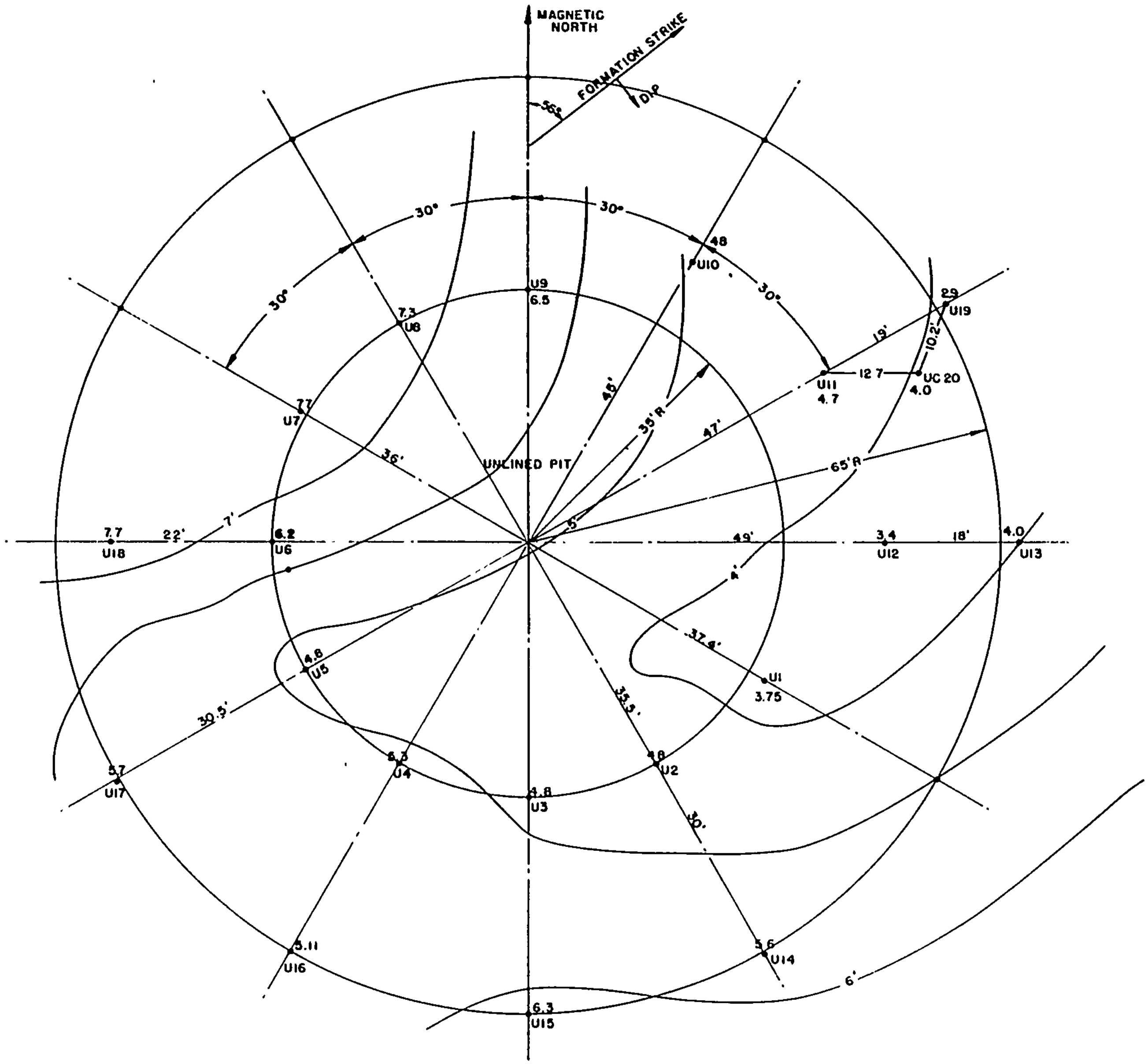

LEGEND
U1• OBSERVATION AND SAMPLE WELLS (RESULTS OF 5/19/54)
—4— RELATIVE WATER TABLE CONTOURS TO COMMON BASE

Fig. 8B. Unlined Experimental Pit. Water Table Configuration.

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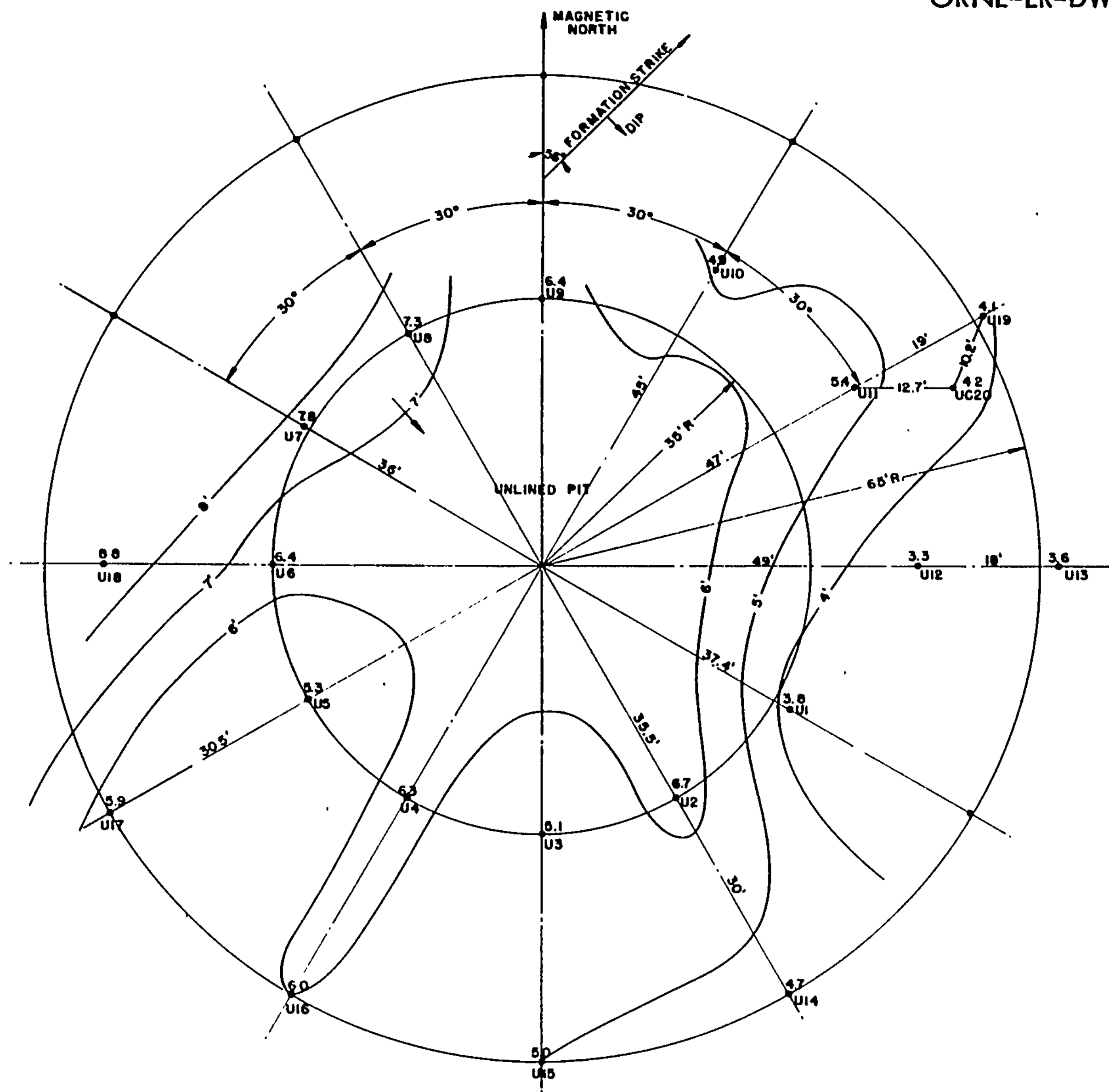

LEGEND

U1• OBSERVATION AND SAMPLING WELLS (RESULTS OF 6/2/54)

4 RELATIVE WATER TABLE CONTOURS TO COMMON BASE.

Fig. 8C. Unlined Experimental Pit. Water Table Configuration.

Analysis of the data obtained implied that the pit was leaking at a mean rate of about 0.02 ± 0.03 gal per sq ft per day; amounting to a mean loss of 8.3 ± 13.1 gal per day. The sprayed asphalt liner, as placed in the field, was more permeable by a factor of about 10^3 as compared with similar liner materials investigated in the laboratory. Non-uniformity of the asphalt liner thickness and puncturing of the liner with tamped shale are deemed responsible for the increase in permeability.

Core holes were drilled to define the geology of the area and specific study was begun of a four-acre site selected for high level waste disposal. Four 4-in diameter core holes (200 ft deep) and a 6-in diameter churn hole (300 ft deep) were drilled and readied for recharge and pumping tests by the USGS to determine the permeability of the soil and rock formations and movement of ground water in this area. Pertinent data will be assembled and suitable geological and hydrological maps of the four-acre site and other parts of the ORNL area will be prepared.

ORNL and Off-Site Wastes. Pilot scale units, consisting of three up-flow sludge contact basins and ion exchange units, were evaluated for the removal of radioactive materials from the large volume-low level process wastes discharging from the ORNL Settling Basin. This study was a joint ERDL-PHS-ORNL endeavor. The results of the investigation were submitted to the ORNL Operations Division(10).

A third chemical waste pit was constructed and placed in operation, because ORNL chemical waste pit #2 had reached its capacity. Chemical waste pits 1 and 2 received highly alkaline wastes containing radioactive cesium and ruthenium. Waste pit #3 will receive a similar waste, but it will be more dilute by a factor of about 10.

Correlated Activities. Included here are cooperative studies with the USPHS, ERDL, TVA, and USGS.

Ceramics Study

The program of investigation of the Ceramics Group, Metallurgy Division, ORNL, in support of the liquid waste studies, included the physical characteristics of clays and other materials for use as mineral liner materials, of gel producing properties of waste solutions with limestones and dolomites and the use of these gels as liner materials in conjunction with waste pits for acid waste materials, and the production of low temperature (1800° F or less) ceramic masses to retain and fix radioactive materials.

The use of Tennessee ball clay as a liner material for use with waste pits receiving neutral or alkaline wastes has been evaluated in the laboratory (Figure 9) and confirmed on a semi-pilot plant scale by the Sanitary Engineering Research Section, as indicated earlier. The gels produced by limestones and dolomite in contact with acid waste solutions formed liners which were quite resistant to the passage of radioactive strontium (Figure 10). Semi-pilot scale confirmation of these laboratory studies is under way. Preliminary studies have shown that ceramic bodies can be prepared with Conasauga shale and the acid waste solution in the presence of fluxing agents. These studies have shown that strontium is fixed in the ceramic lattice and is not leached out by tap water. Further investigation using other radioisotopes and leaching solutions will be made.

The major emphasis of the Ceramics program will be on the feasibility of utilizing the inherent heat from the radioactive decay of the fission products to fuse and fix the activity on Conasauga shale.

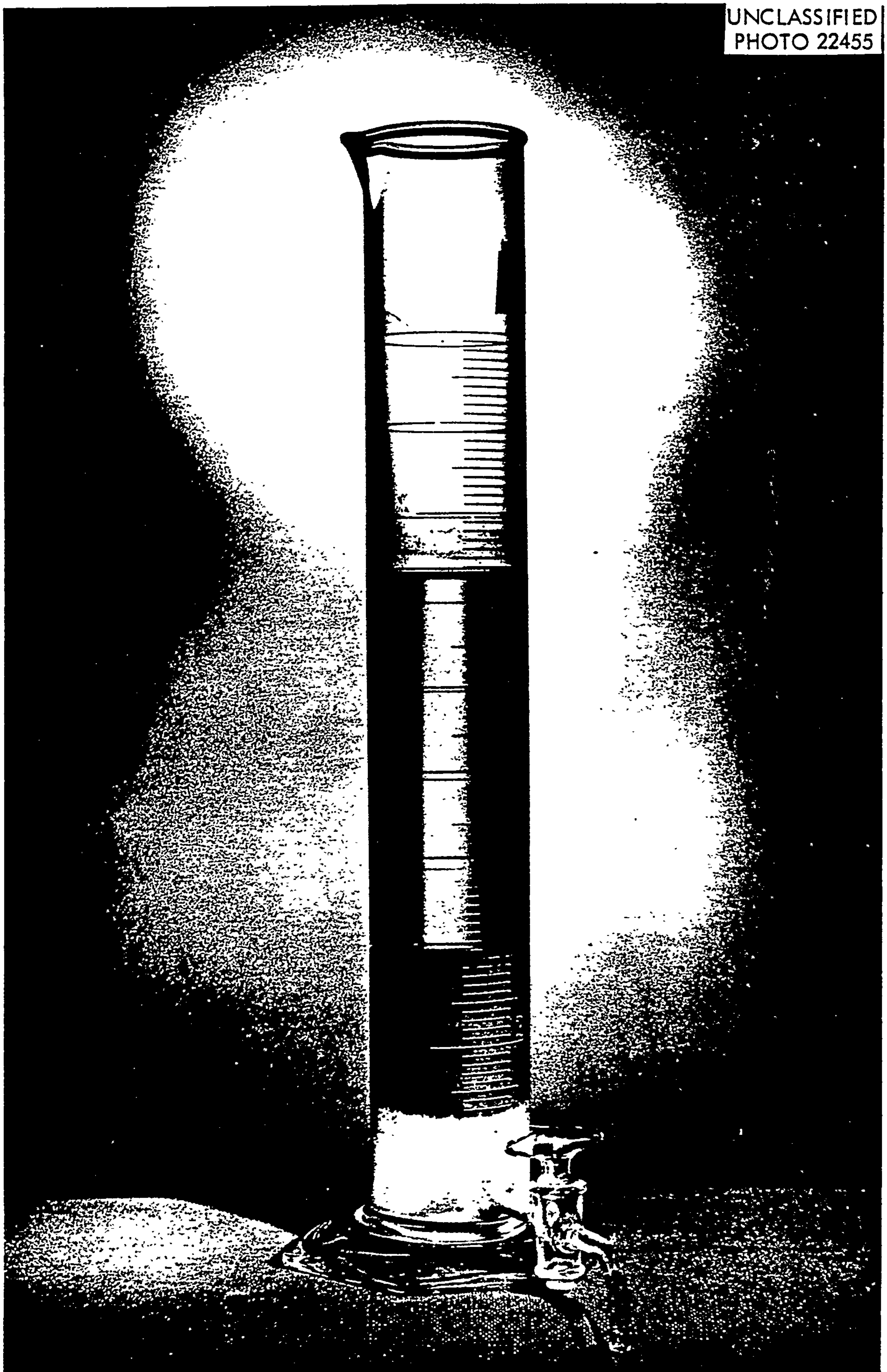

Fig. 9. Graduated Cylinder Used to Evaluate Mineral Liners on Laboratory Scale.

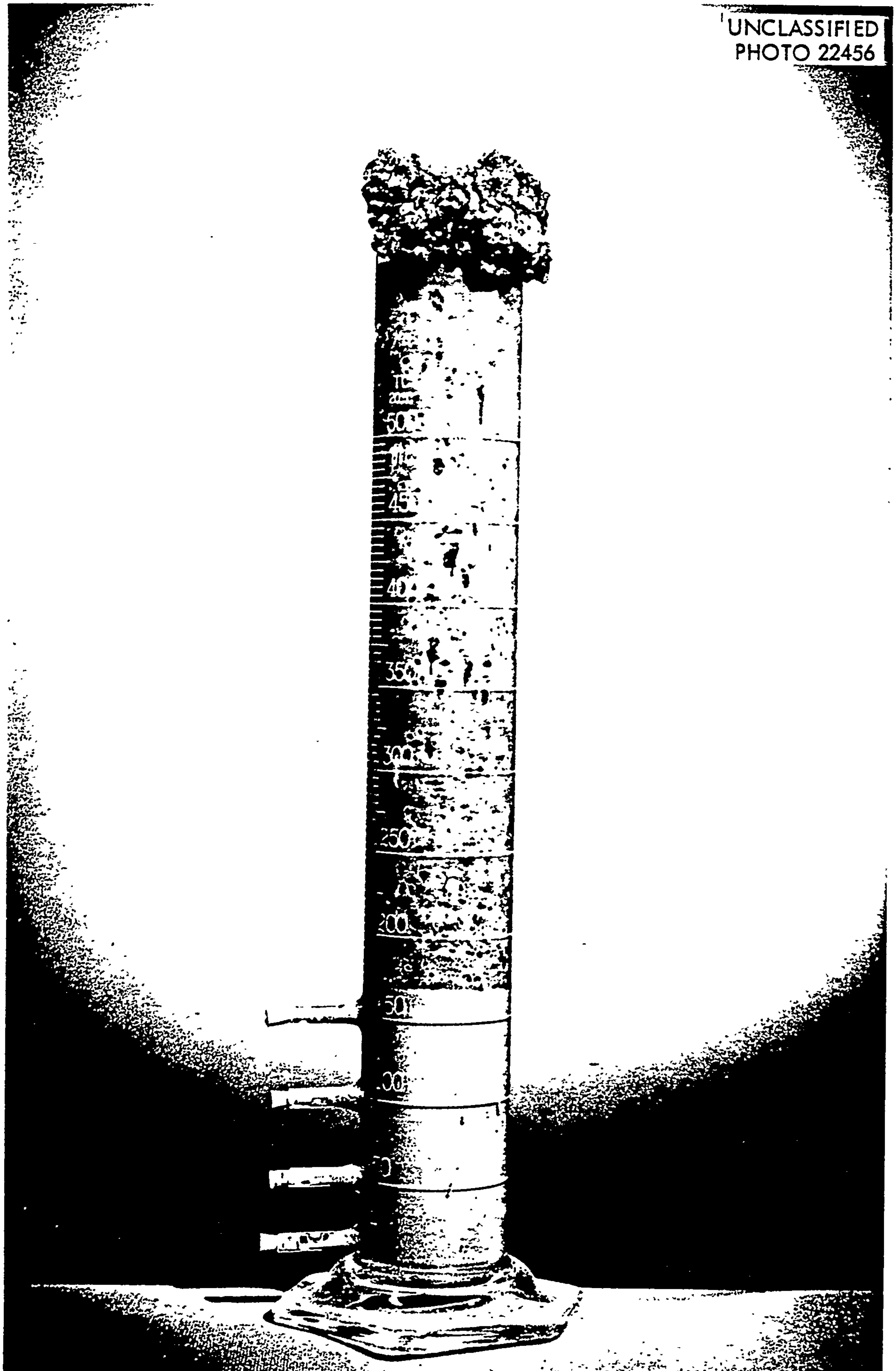

Fig. 10. Apparatus for Laboratory Evaluation of Gels.

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